NASA CR-54272

Development of

HIGH-TEMPERATURE, HIGH-CURRENT, ALKALI-METAL, VAPOR-FILLED CERAMIC THYRATRONS AND RECTIFIERS

by

A. W. Coolidge

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

CONTRACT NAS3-6005

GENERAL ELECTRIC

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Quarterly Progress Report No. 2

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January 6, 1965

CONTRACT NAS3-6005

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Schenectady, New York 12305

TABLE OF CONTENTS

LIST OF ILLUSTRATIONS

Development of
HIGH-TEMPERATURE, HIGH-CURRENT, ALKALI-METAL
VAPOR-FILLED CERAMIC THYRATRONS AND RECTIFIERS

by A. W. Coolidge

General Electric Company

SUMMARY

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The purpose of the National Aeronautics and Space Administration Contract NAS3-6005 is to advance the technology and to provide fundamental design data for high-temperature, high-current, alkali-metal, vapor-type ceramic tubes. The objectives of this program are to conduct a fundamental investigation of the problem areas associated with high-temperature, vapor-type tubes, and to fabricate and test prototype rectifiers and thyratrons to prove the technology and to provide application data for future reference. Phase I of this program is concerned with the investigation of the fundamental problems and the establishment of the conceptual design of prototype models.

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During the report period, a meeting was held between the National Aeronautics and Space Administration and the General Electric Company in which it was agreed that the reservoir portion of the tube could radiate to a heat sink of approximately 300°C. The reduction of the environmental temperature of the radiator from 600°C will permit the study of pure cesium as a filling agent for tubes to be made in accordance with the contract requirements.

The following work on alkali halides which was in progress at the time of the meeting, was continued:

1. Nickel-plated ceramic seal structures were made
2. A seal structure was made using a palladium-cobalt brazing compound
3. Seal structures were made using a brazing compound high in nickel and low in titanium content.

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ABST Continued

During the reporting period, effort was also devoted to an evaluation of the inverse current from a molybdenum anode, and to an analysis of the effect of work function on spurious emission from an anode or grid.

In addition, the test diode was redesigned to increase its inverse voltage ability and to decrease leakage between electrodes.

Finally, construction was started on two tubes. These tubes will be filled with thallium and will have barium system cathodes.

AUTHOR

INTRODUCTION

Two cesium-filled devices were used to demonstrate that low tube-drop operation could be achieved. In fact, the demonstration showed that the tube drop could be made negative by elevating the cathode temperature to 1450°C.

It is recognized that there are more variables involved in designing an alkali-halide tube than in designing a tube filled with cesium only. Early in the report period, a meeting was held at the National Aeronautics and Space Administration. At this meeting, NASA personnel directed that (at least for the near future) the major effort on the contract be shifted from alkali-halide to cesium-filled devices.

Inverse-current and voltage measurements, taken on early cesium diodes, indicated a ceiling voltage of about 150 volts and excessive back emission at an anode temperature of 500°C or above. As a result, in a redesign of the test diode, the spacings were adjusted to optimize the voltage capability. The effect of work function on spurious grid or anode emission was studied. A few anodes were prepared and in order to reduce spurious emission they were coated with materials having promising work functions.

During this report period, some additional work was performed with CsI. This work was done in an attempt to reduce a troublesome halogen cycle that had occurred in earlier tubes.

Thallium-filled devices are under construction. When completed the characteristics of these units and cesium-filled devices will be compared.

A conceptual design has been delineated for a high-temperature thyratron.

PUBLICATIONS, LECTURES, REPORTS AND CONFERENCES

PUBLICATIONS - None

LECTURES - None

REPORTS

1. Monthly Progress Report No. 3
   Period Covered: September 19, 1964 through October 18, 1964
   Author: A. W. Coolidge

2. Monthly Progress Report No. 4
   Period Covered: October 19, 1964 through November 18, 1964
   Author: A. W. Coolidge

CONFERENCES

1. Organizations represented:
   NASA
   E. A. Koutnik
   H. A. Shumaker
   R. L. Cummings
   R. English
   J. Ward
   Dr. B. Lubarsky
   L. K. Tower
   H. F. Butze
   Missile Space Division, General Electric Company
   R. Edwards
   Schenectady Tube Operation, General Electric Company
   E. A. Baum
   A. W. Coolidge
   A. Michaelson
   Defense Products Operation, General Electric Company
   M. Toth
   Place and date: NASA Lewis Research Center, Cleveland, Ohio
   October 19, 1964
   Subject: Review technical progress on this project.

TECHNICAL DISCUSSION AND PROGRESS

LOW VOLTAGE-DROP OPERATION

Two tubes with pure cesium fill, designated Z-7009 Nos. 2 and 3, were made according to Design A shown in Figure 1. Tube 2 had an anode-to-cathode spacing of 0.030 inch, while tube 3 had an anode-to-cathode spacing of 0.010 inch.

When these tubes were operated at currents up to 15 amperes average, extremely low arc drops were observed. Indeed, at low average currents and high cathode temperature, the tube drop was actually negative, indicating that the tube was capable of "generating" like a thermionic converter.

The operating data on these tubes is summarized in Figures 2, 3, 4 and 5. As can be seen from these illustrations, the closest spaced tube exhibited the lowest drop. Under pulse conditions, the tube was loaded with a current of 104 amperes, equivalent to about 20 amperes per square centimeter, with a tube drop of 1.6 volts. In order to obtain high-density cesium-enhanced cathode emission, it is necessary that the cesium pressure, which determines the arrival rate of cesium atoms at the cathode surface, be relatively high - in the order of 0.5 to 1 Torr. At such a pressure, the maximum voltage that may be impressed across a tube without cold breakdown is limited to a relatively low value. Because the voltage level decreases as the product of pressure and distance increases, maximum voltage capabiltity is realized by keeping interelectrode gap dimensions to a small value.

GRID EMISSION AND INVERSE EMISSION

The ability of a thyratron to hold off high anode voltages depends upon the grid emission being suppressed to a low value. Likewise, if the tube is to withstand high inverse voltages, reverse current caused by emission from the anode must be negligible. Cold breakdown will occur when the voltage exceeds the "PD" or Paschen limitation, or breakdown may occur as a result of the thermionic emission (from grid or anode) reaching such a magnitude that the collision process causes ionization.

Figure 6 shows inverse-current measurements taken on the Z-7009 tube 2 which has a molybdenum anode surface. As shown in this illustration,

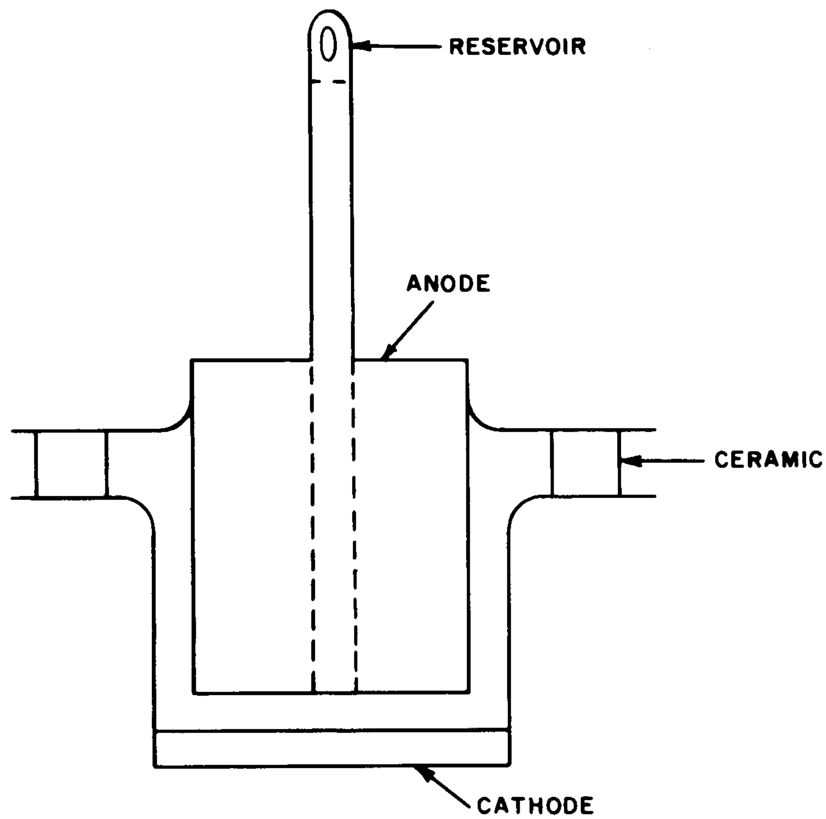

Figure 1 - Schematic of Test Vehicle, Design A

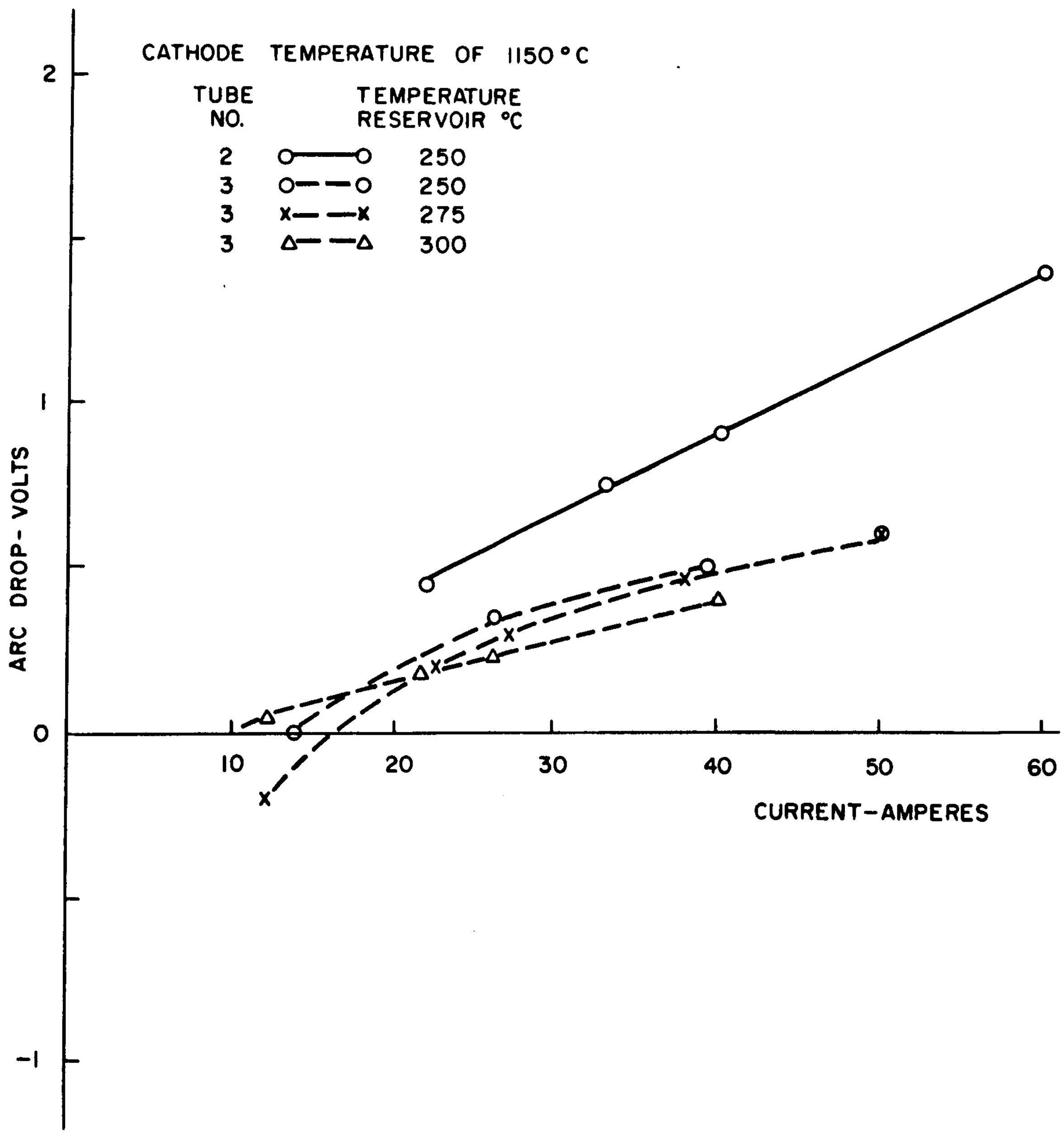

Figure 2 - Peak Tube Drop versus Peak Current for Z-7009 Tubes 2 and 3, Cathode Temperature 1150°C

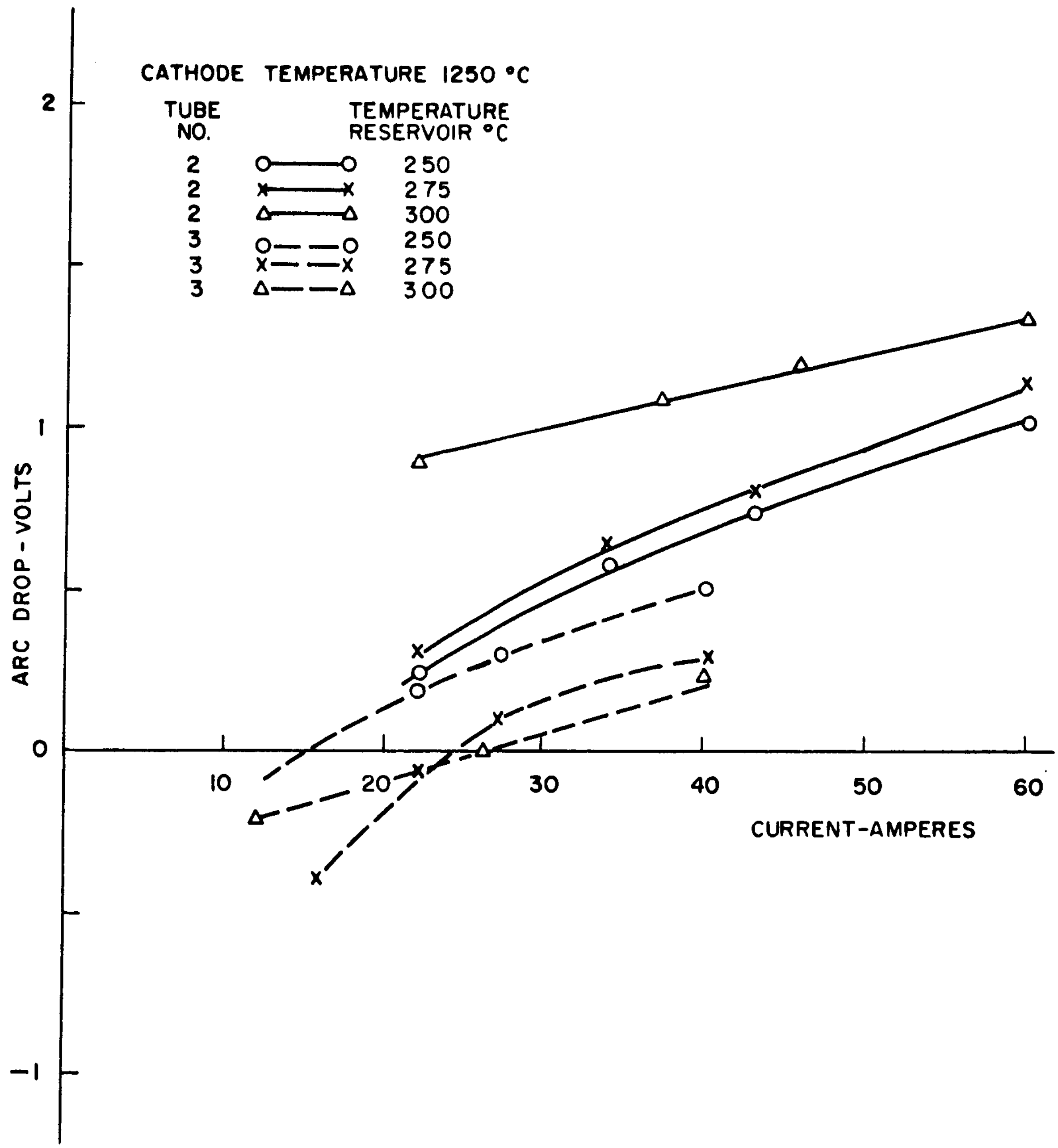

Figure 3 - Peak Tube Drop versus Peak Current for Z-7009
Tubes 2 and 3, Cathode Temperature 1250°C

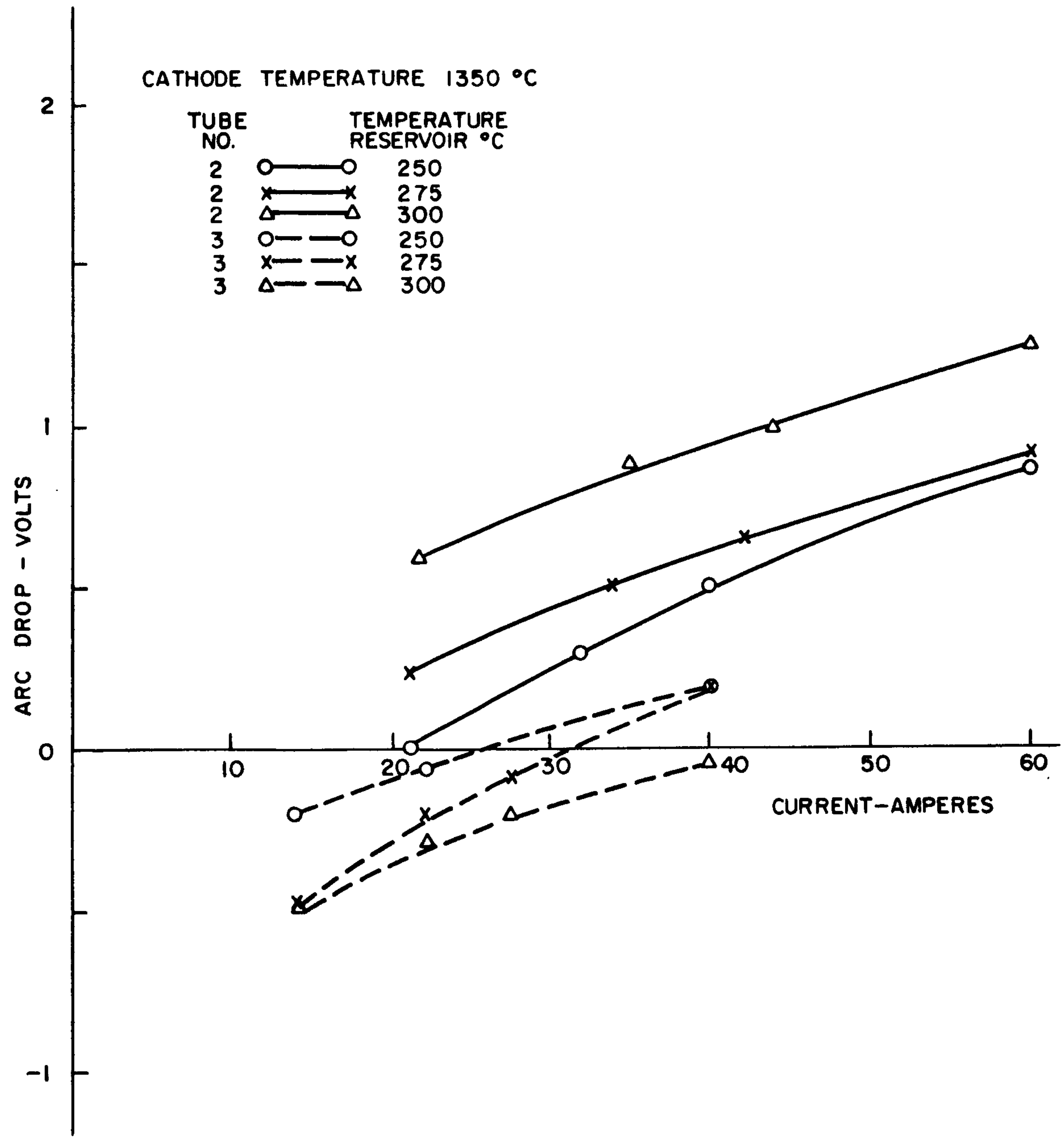

Figure 4 - Peak Tube Drop versus Peak Current for Z-7009 Tubes 2 and 3, Cathode Temperature 1350°C

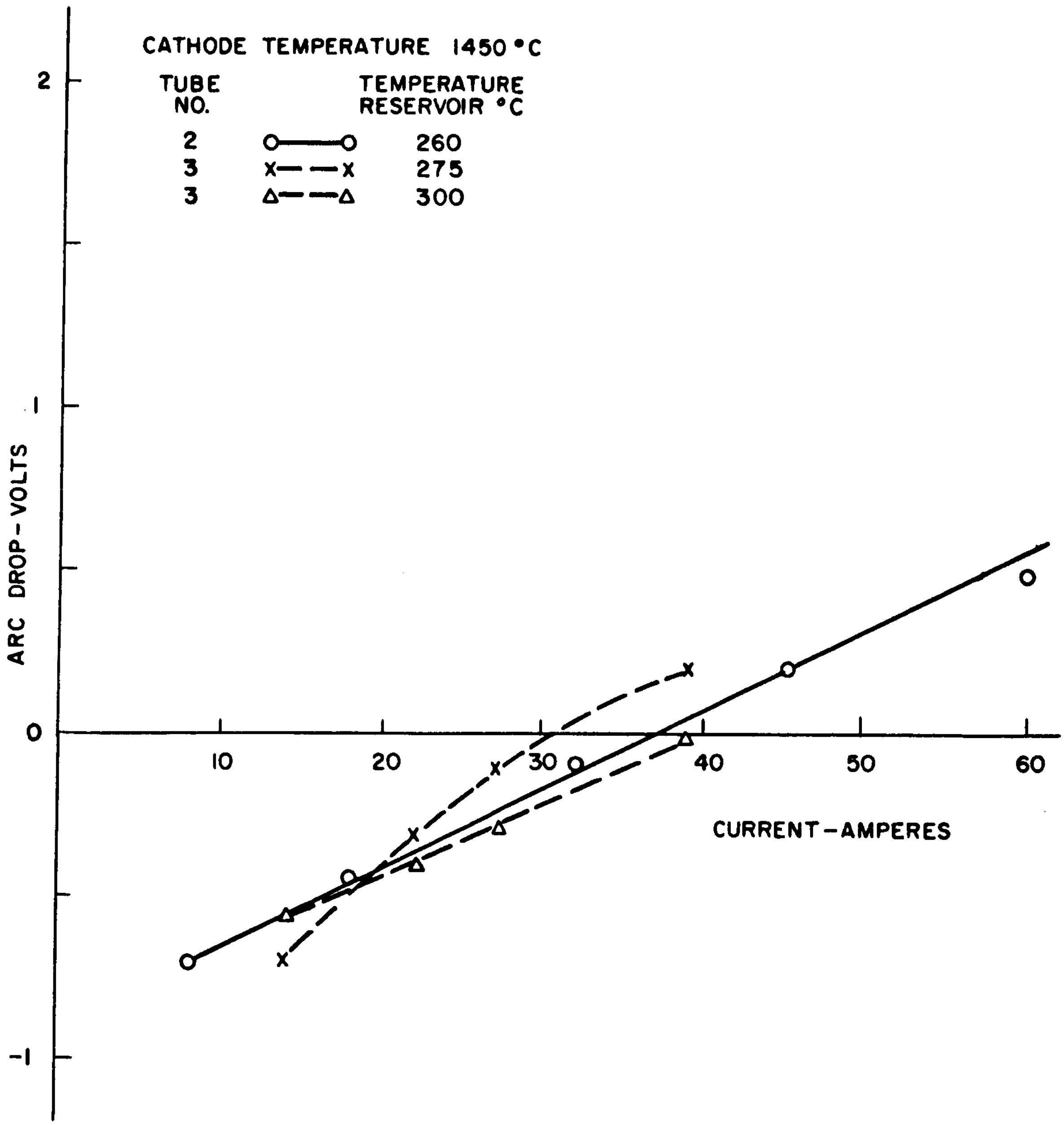

Figure 5 - Peak Tube Drop versus Peak Current for Z-7009 Tubes 2 and 3, Cathode Temperature 1450°C

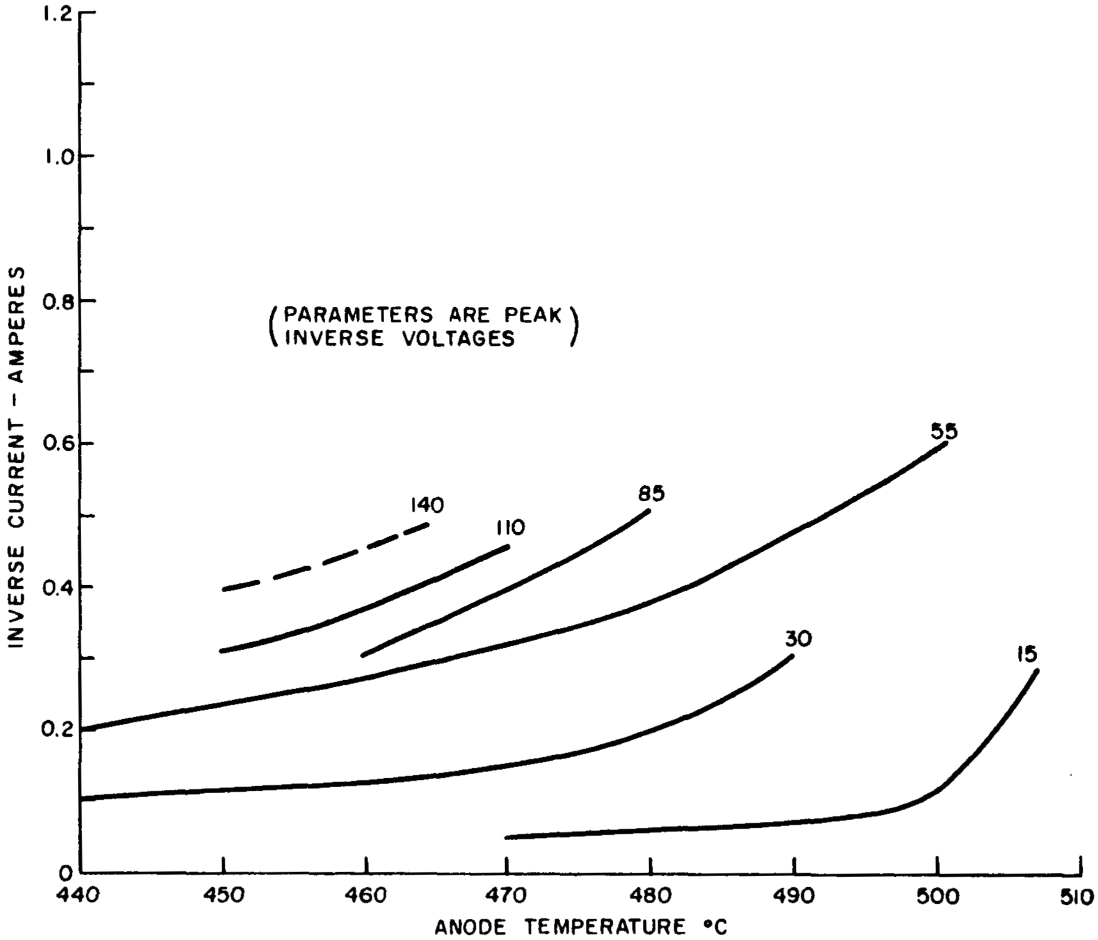

Figure 6 - Inverse Current versus Anode Temperature as a Function of Inverse Voltage

the inverse current was excessive when the anode temperature approached 500°C. The failure of the curves to approach zero current at low temperature indicates that the tube suffered from a leakage path of about 300 ohms. The data is limited to peak inverse voltages of 140 volts, because inverse breakdown occurred at 150 volts. Assuming a temperature drop of about 200°C, between a heat sink external to the tube and the hottest part of an electrode connected to the heat sink, an arbitrary temperature rating of 125 volts inverse and 250°C ambient might be applied to this particular diode. These values are suggested merely to point out the amount of improvement that must be realized in order to achieve the objectives of 750 volts inverse and 600°C ambient temperature.

The design of the Z-7009 tube 2 is essentially that of a thermionic converter, and it was built primarily for studying cesiated cathode emission characteristics. In this tube, voltage breakdown occurred across the insulator which was the longest interelectrode path within the tube. In a redesign of the test vehicle (Figure 7), two changes were made:

1. The ceramic insulator was increased in length from 0.090 to 0.50 inch to reduce or eliminate leakage path difficulties

2. All interlectrode spacings, except in the planar gap area, were reduced to about 0.030 inch; the size of the planar gap was left at 0.050 inch.

Thus, in this tube cold breakdown should occur only in the region of the planar gap, since the gap constitutes the longest path in the tube. An increase in voltage capability should be realized, since the long path of 0.050 inch in Design B is different from the long path in tube 2 by a factor of two.

It is expected that the geometry in Figure 7 will produce a voltage capability of about 300 volts in a high-pressure cesium tube where the reservoir temperature is 300°C. In a tube with a barium cathode, where a cesium pressure of 100 microns would be adequate, the voltage capability should exceed 1000 volts.

It is of interest to contemplate how <u>poor</u> an emitter an anode (or grid) surface must be to prevent spurious ionization. One estimate can be made by computing the saturation current realized from cesiated molybdenum at 500°C - these conditions corresponding to the run-away threshold of the Z-7009 tube 2. Although the work function of bare molybdenum is about 4.4, its work function when cesiated drops to about 2.0 (as indicated in a later

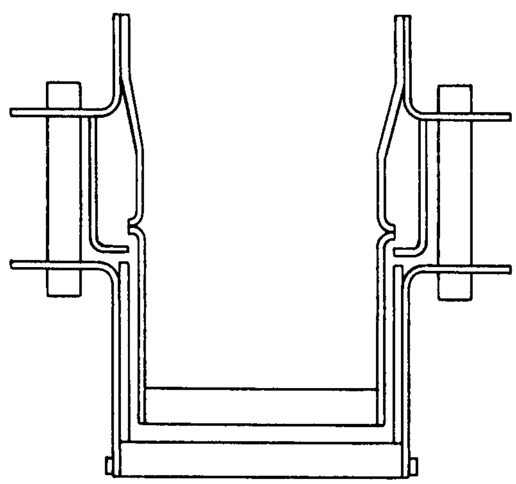

Figure 7 - Schematic of Test Vehicle, Design B

section, entitled "Effect of Work Functions" and shown in Figure 10). The curves in Figure 8, drawn for Richardson's equation, indicate a saturation current of about 10^{-5} ampere per square centimeter.

Another independent check, although somewhat crude, was made with a GL-6807 industrial thyratron, Figure 9. In this experiment, 750 volts, DC forward was applied to the grid of the tube while the cathode was at room temperature. At such a low temperature, the barium-composition cathode, notwithstanding its low work function of about 1.5, did not yield enough electron current to ionize the grid-cathode region. The cathode was gradually heated until breakdown of this region did occur, at a temperature of about 330°C. Again, the saturation current obtained from Figure 8 was 10^{-5} ampere per square centimeter. While the actual area of the cathode is 50 square centimeters, in this experiment only a small portion at the top of the cathode structure and nearest the grid would contribute electron current prior to breakdown. It was assumed that the effective cathode area was about one square centimeter.

The two preceding examples indicate that emission from anode or grid should not exceed approximately one microampere per square centimeter in order to prevent spurious ionization or breakdown in the tube. This estimate should help in selecting grid and anode materials for a high-temperature thyratron.

EFFECT OF WORK FUNCTIONS

When a substrate material is coated with a thin layer of another material, the work function of the combination is generally different from the work functions of either of the individual materials. For example, cathode emission is enhanced by a layer of barium molecules on nickel, or by a layer of thorium on tungsten. Indeed, when a monolayer of cesium resides on a refractory metal, such as molybdenum or tungsten, both of which have work functions higher than 4.0, the work function of the combination is reduced to a value of 2.0 or lower.

Spurious emission from a grid or anode in a cesium tube cannot be predicted until a relationship is established between the substrate work function and the cesiated work function over the range of interest.

Rasor and Warner[1] have attempted to establish just such a relationship.

1. Rasor, N. S., Warner, C., Correlation of Emission Processes for Adsorbed Alkali Films on Metal Surfaces, Journal of Applied Physics, Volume 35, Number 9, September 1964, pp. 2589-2600.

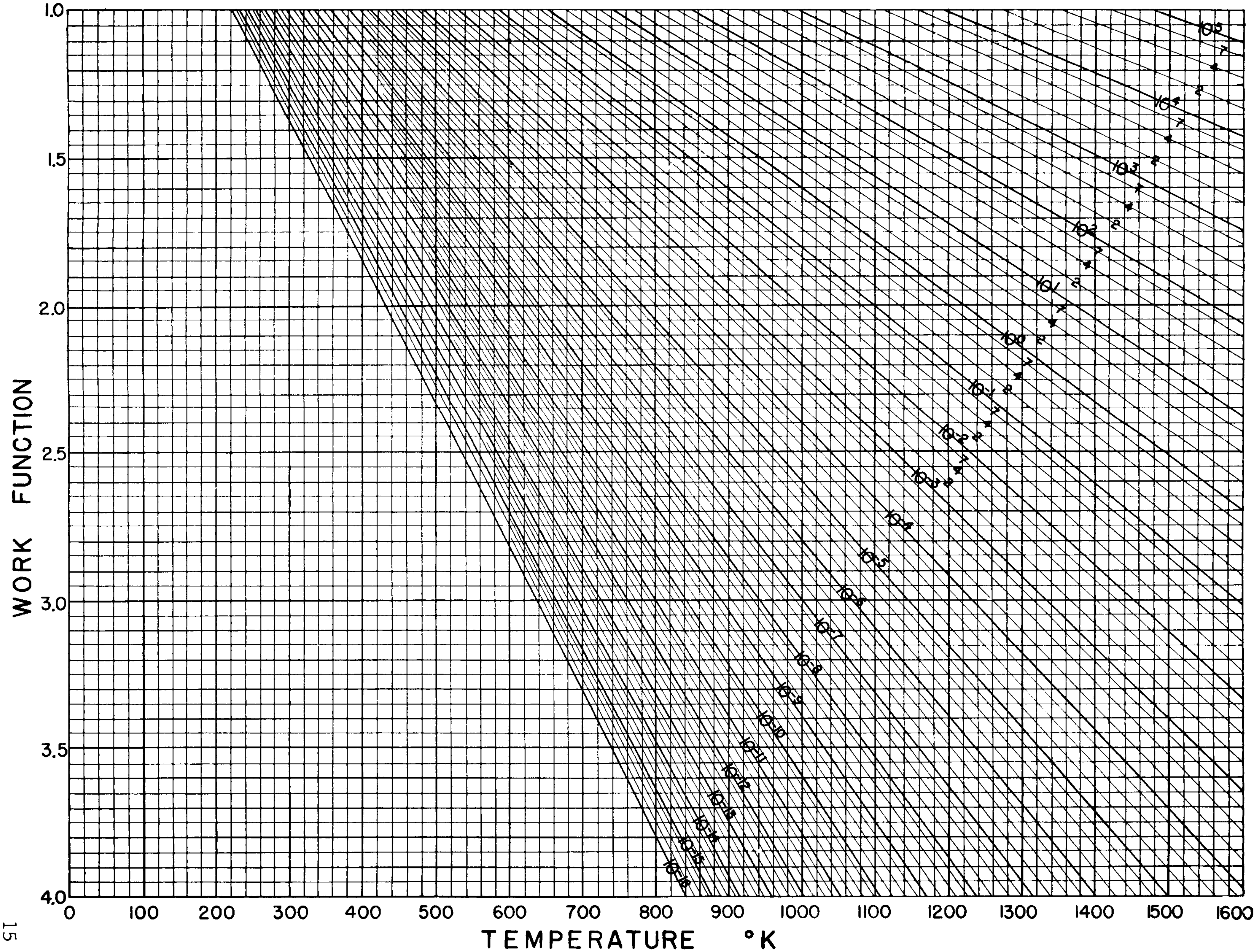

Figure 8 - Relation Between Work Function, Temperature and Saturation Emission (Richardson's Equation)

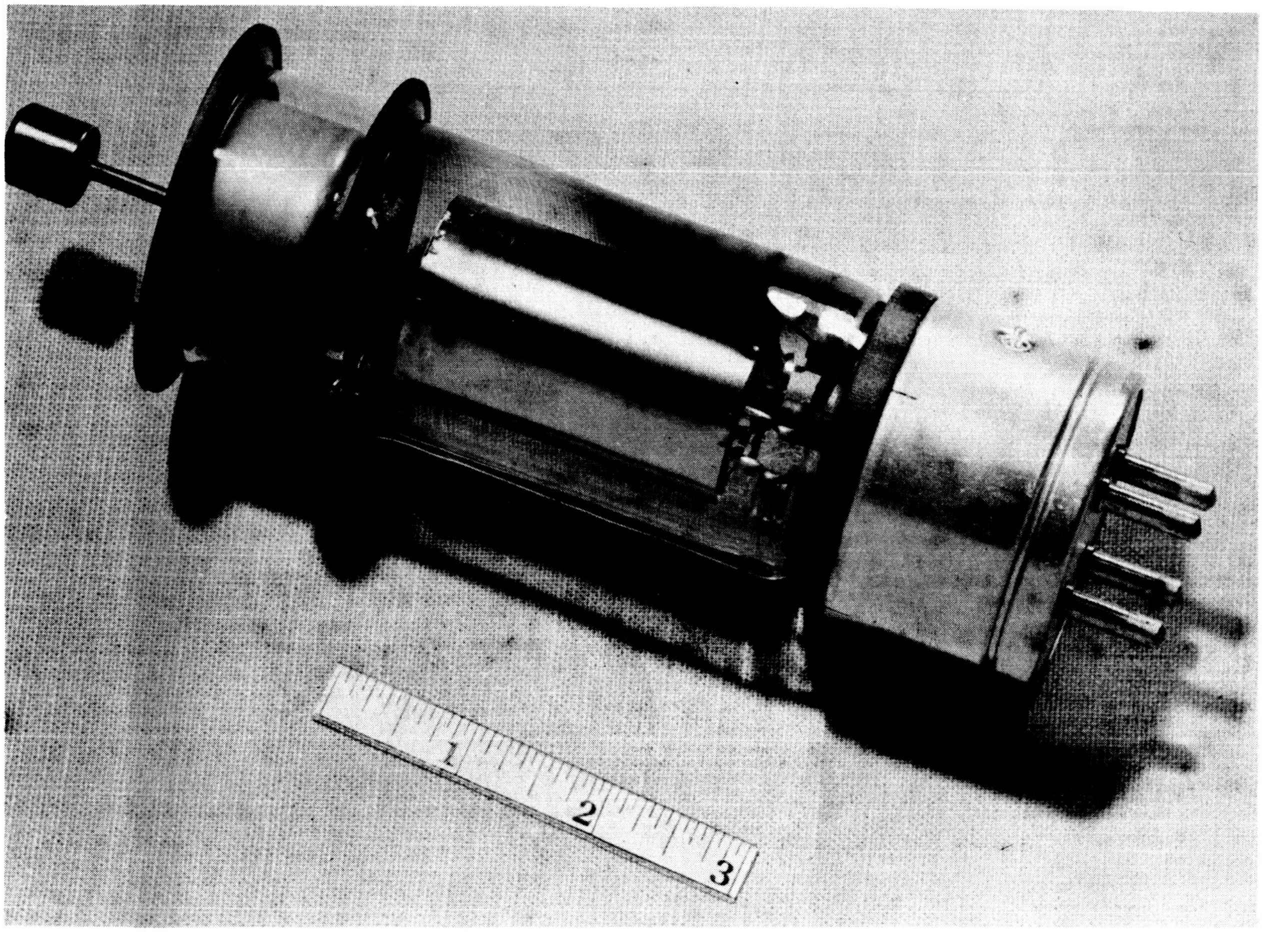

Figure 9 - GL-6807 Industrial Thyratron

Figure 10 displays a family of curves computed for this relationship. Although the authors point out that some parts of the curves are subject to questionable validity, they are helpful in attacking the problem of spurious grid and anode emission. Note in Figure 10 that for a low substrate work function, the cesiated work function remains the same, but as the substrate work function increases through the range of 3.0 to 5.0, the cesiated work function becomes lower. Therefore, the highest cesiated work function corresponds to a substrate work function of midvalue, about 3.0 to 3.5. By combining the information given in Figures 8 and 10, a new series of curves may be drawn. These curves (Figures 11, 12, 13 and 14) give the relationship between saturation current and substrate work function. For convenience, the same data is presented in Figure 15 in terms of cesiated emission versus bare substrate work function. In this illustration, the range of surface temperatures will cover grid and anode temperatures of the objective high-temperature thyratron. With high thermal conductivity coupling between tube elements and a 600°C heat sink, the surface temperature of anode and grid would be approximately 800°C.

From Figure 15 it is obvious that a work function of 3.0 would be preferable for the anode and grid surfaces. While a substrate work function of 6.0 would also be fairly good (for low emission), few conductors have such a high work function. Platinum has a work function range of 4.7 to 6.3, depending upon the exact crystallographic arrangement at its surface. However, platinum does not seem dependable, since if its work function slipped to 4.7 it would become an excellent emitter.

Two materials chosen for early evaluation are the compound zirconium carbide and the element hafnium which have work functions of 3.1 and 3.5, respectively. While zirconium carbide has the preferable work function, it is subject to some degree of dissociation which would undoubtedly cause a lower than predicted cesiated work function. On the other hand, hafnium cannot suffer from dissociation so that the actual cesiated work function should be fairly well in line with the predicted value.

Two tubes are under construction (Figure 1) in which the molybdenum anodes have been coated (by a plasma jet spraying process) with zirconium carbide and hafnium, respectively.

ALKALI HALIDES

The first quarterly report described a possible titanium-halogen cycle whereby titanium in the brazing alloy may associate with free halogen (in a tube with alkali-halide fill) and be transported about the tube, later to be

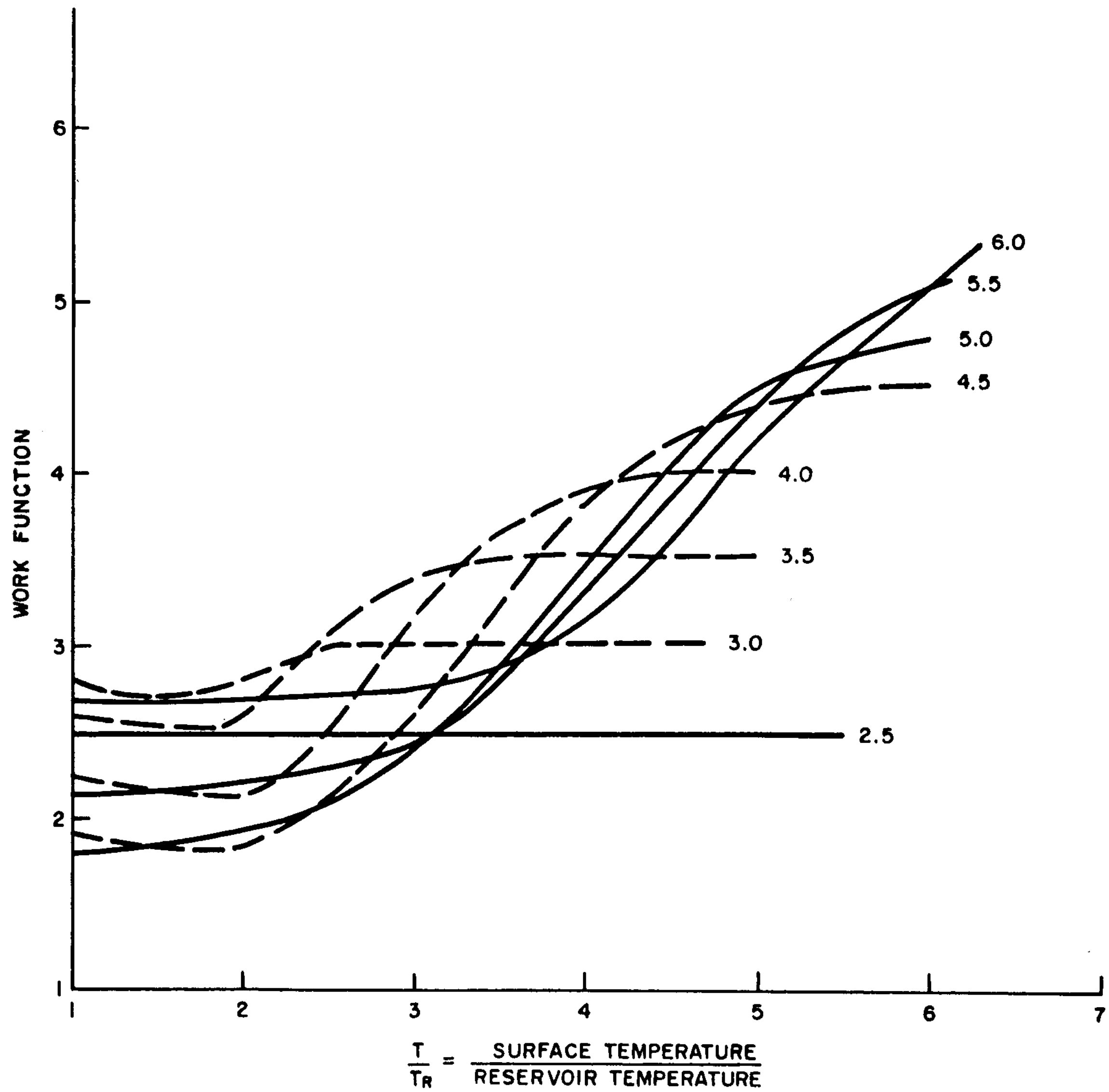

Figure 10 - Cesiated Work Function versus Surface Temperature With Substrate Work Function as Parameter

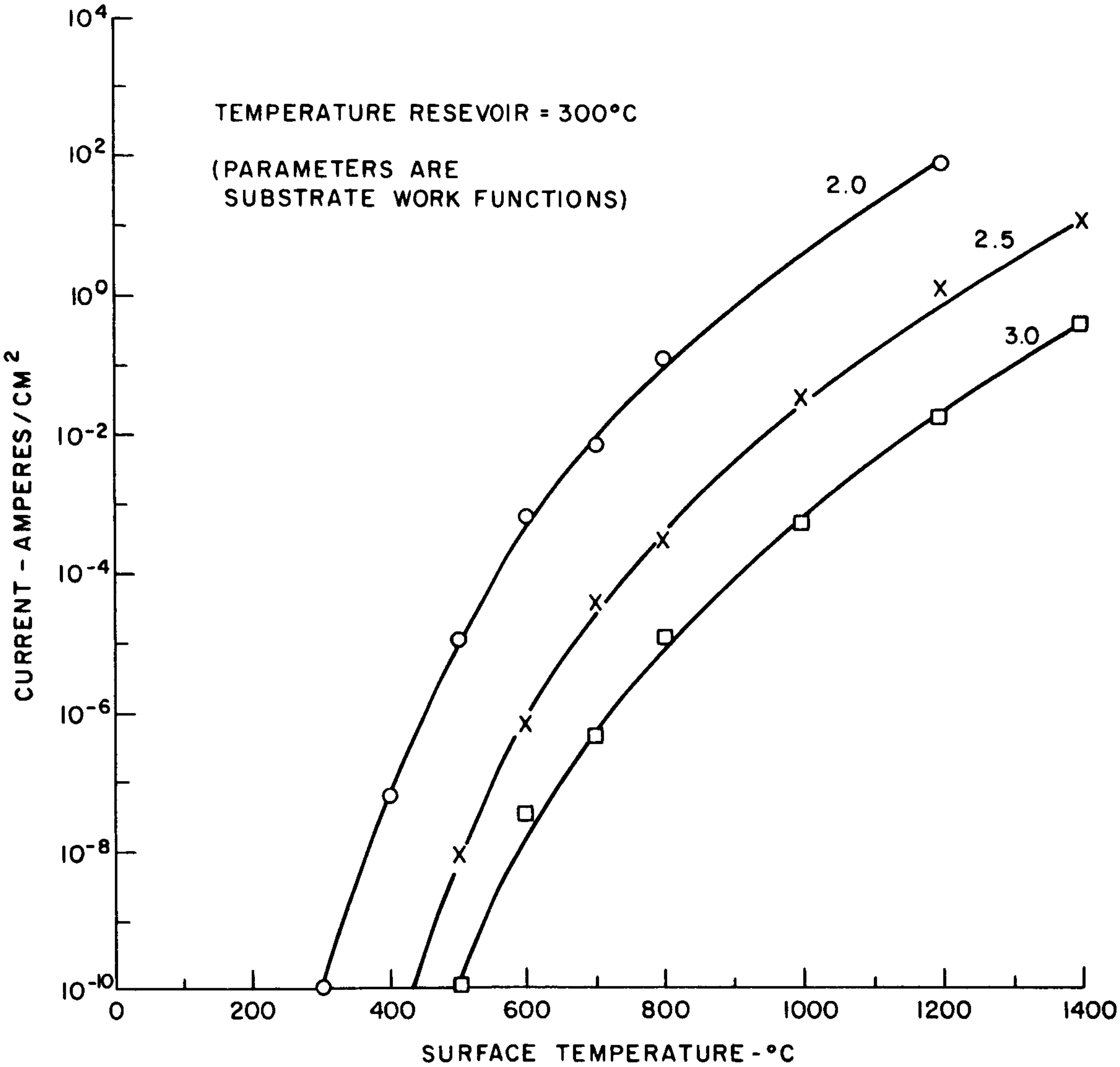

Figure 11 - Cesiated Emission Curves for Low-Range Bare Work Functions

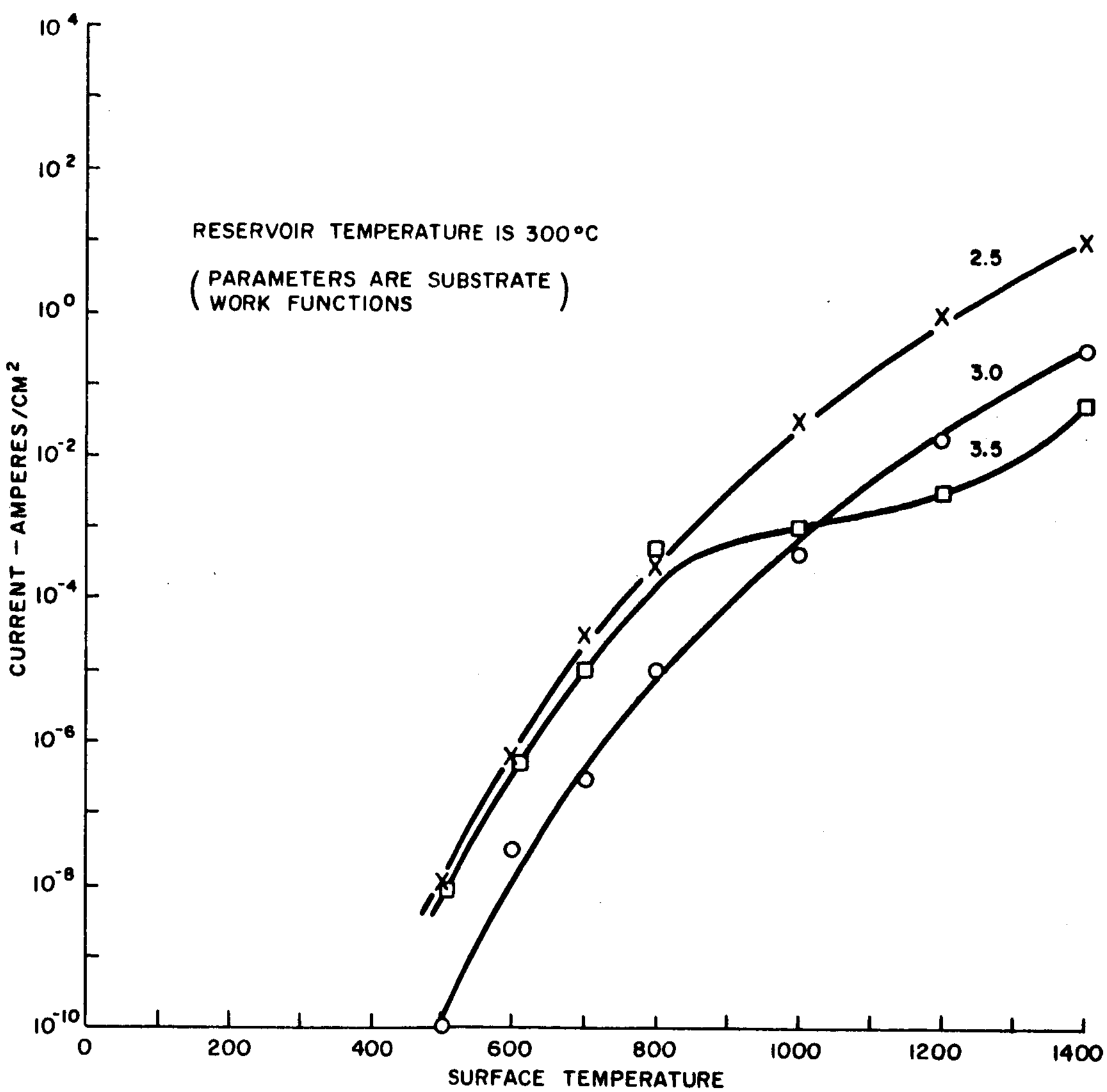

Figure 12 - Cesiated Emission Curves for Middle-Range Bare Work Functions

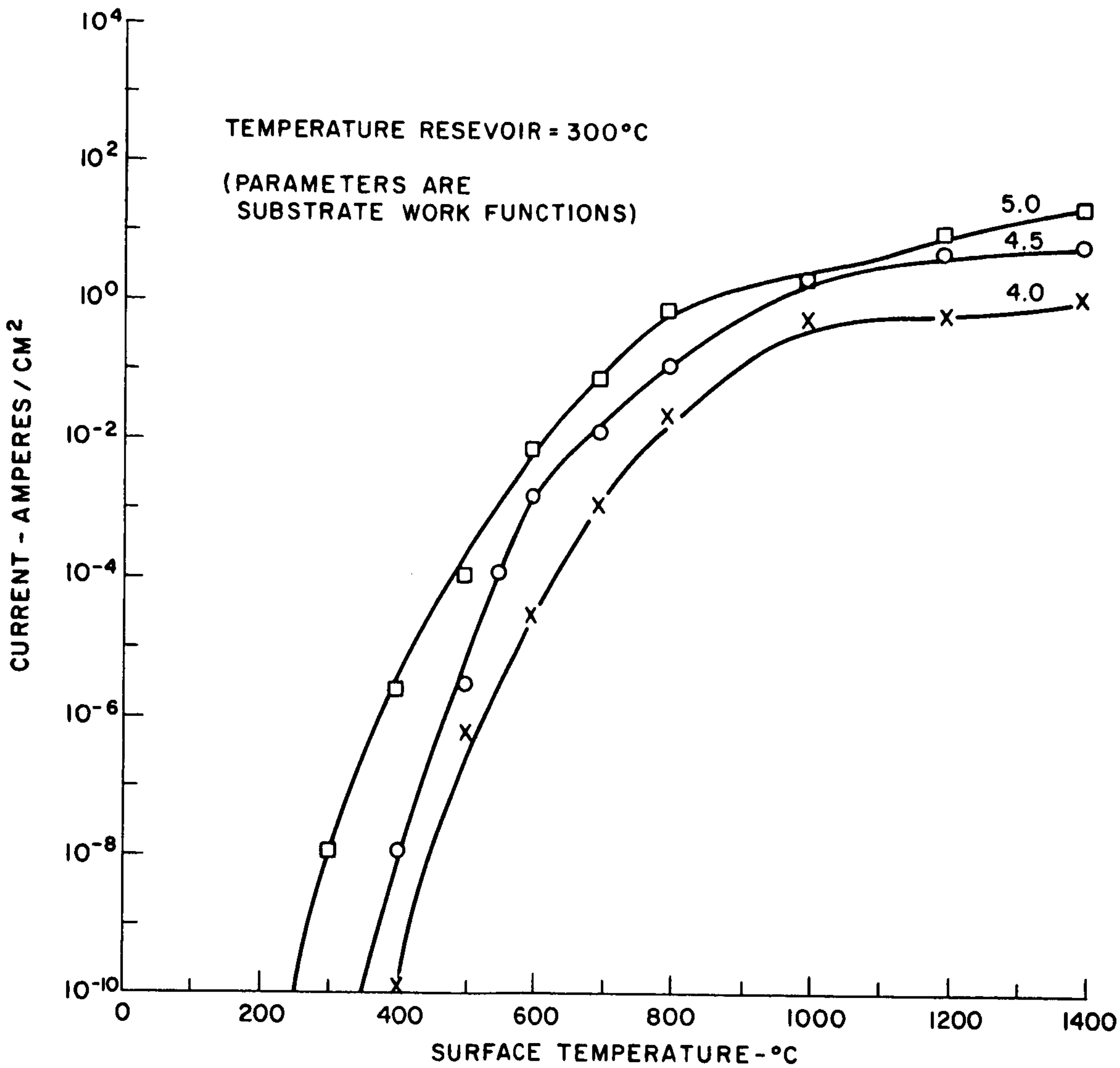

Figure 13 - Cesiated Emission Curves for Middle-to-High-Range Bare Work Functions

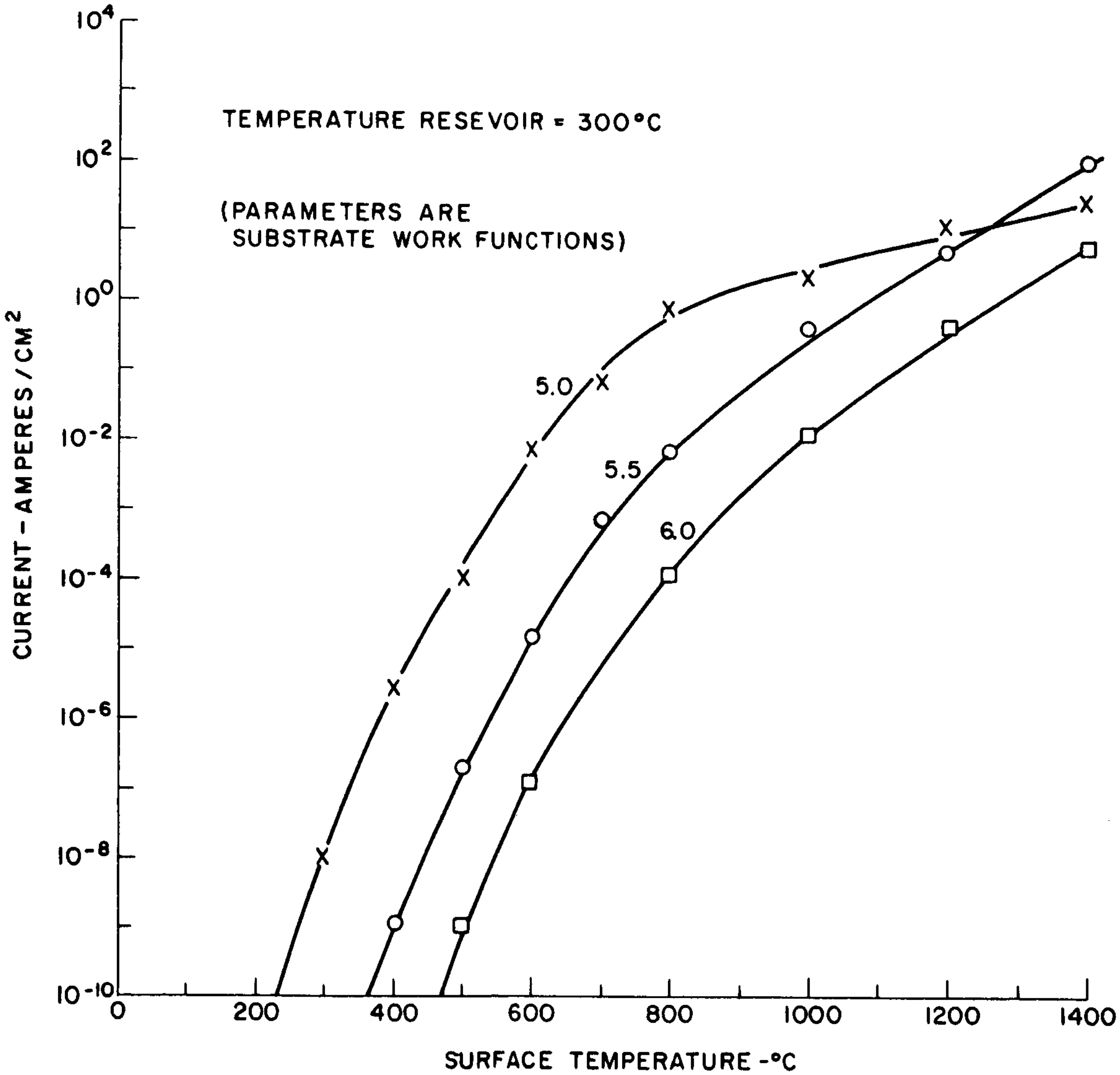

Figure 14 - Cesiated Emission Curves for High-Range Bare Work Functions

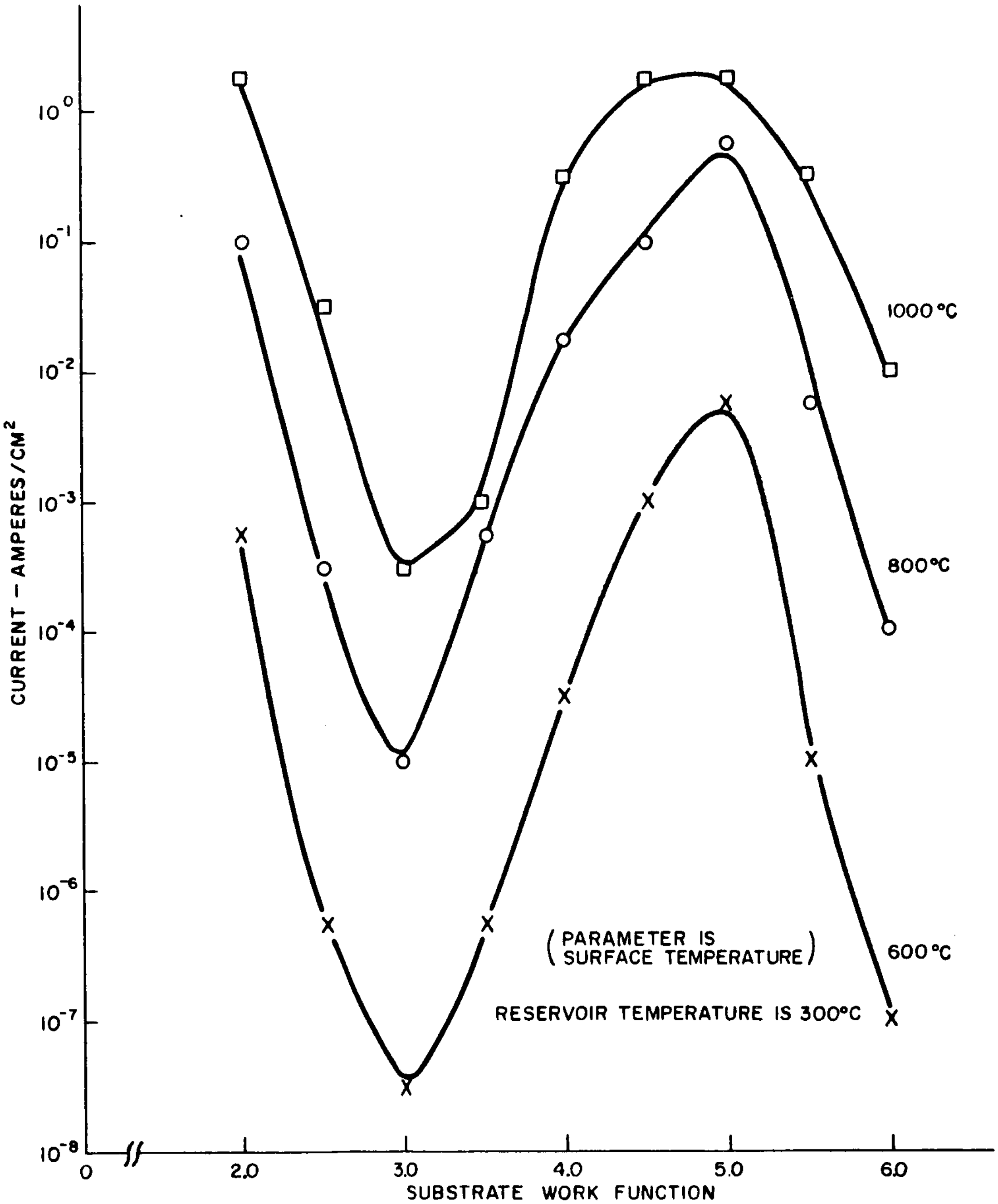

Figure 15 - Cesiated Emission versus Substrate Work Function

deposited and dissociated on some new surface, such as the ceramic separating the tube electrodes. Such a cycle would be consistent with the performance of early tubes in which a coating was laid down on the insulators, causing the tube interelectrode impedance to drop to as low as 0.1 ohm without regard to voltage polarity.

A few projects were under way in the second quarter to combat this cycle by either altering or covering the brazing alloy with an impenetrable coating.

These projects were completed even though emphasis was shifted from alkali-halide fill to pure cesium fill as a result of the October 19 meeting with the technical representatives of NASA. The projects and progress attained on each project are described in the following paragraphs:

1. High-Nickel Low-Titanium Brazing Alloy

The normal titanium-nickel seal is made using a mixture that is 20 percent nickel by weight and which has a eutectic temperature of 942°C, Figure 16.

An attempt was made to change the structure of this alloy, in the hope of minimizing the free titanium available for a halogen cycle, by reversing the ratio of nickel to titanium, producing an alloy having a eutectic temperature of 1304°C.

Although two such seals were made both were leakers, presumably because of the noticeably brittle structure of the high-nickel-content alloy. No further work is planned in this area.

2. Nickel-Plated Seal Areas

It was felt that a halogen cycle might be prevented by covering the conventional titanium-nickel seal with a nickel plate. Accordingly, a seal structure (Figure 17) was designed that would permit the interior to be nickel plated before the cathode and anode were welded into place.

While no particular difficulty was experienced in striking a nickel plate over the seal area within the structure, the first structure became a leaker when the molybdenum cathode was welded into place.

In another structure both the preheating time and temperature were increased before the cathode weld was made. In this structure leak

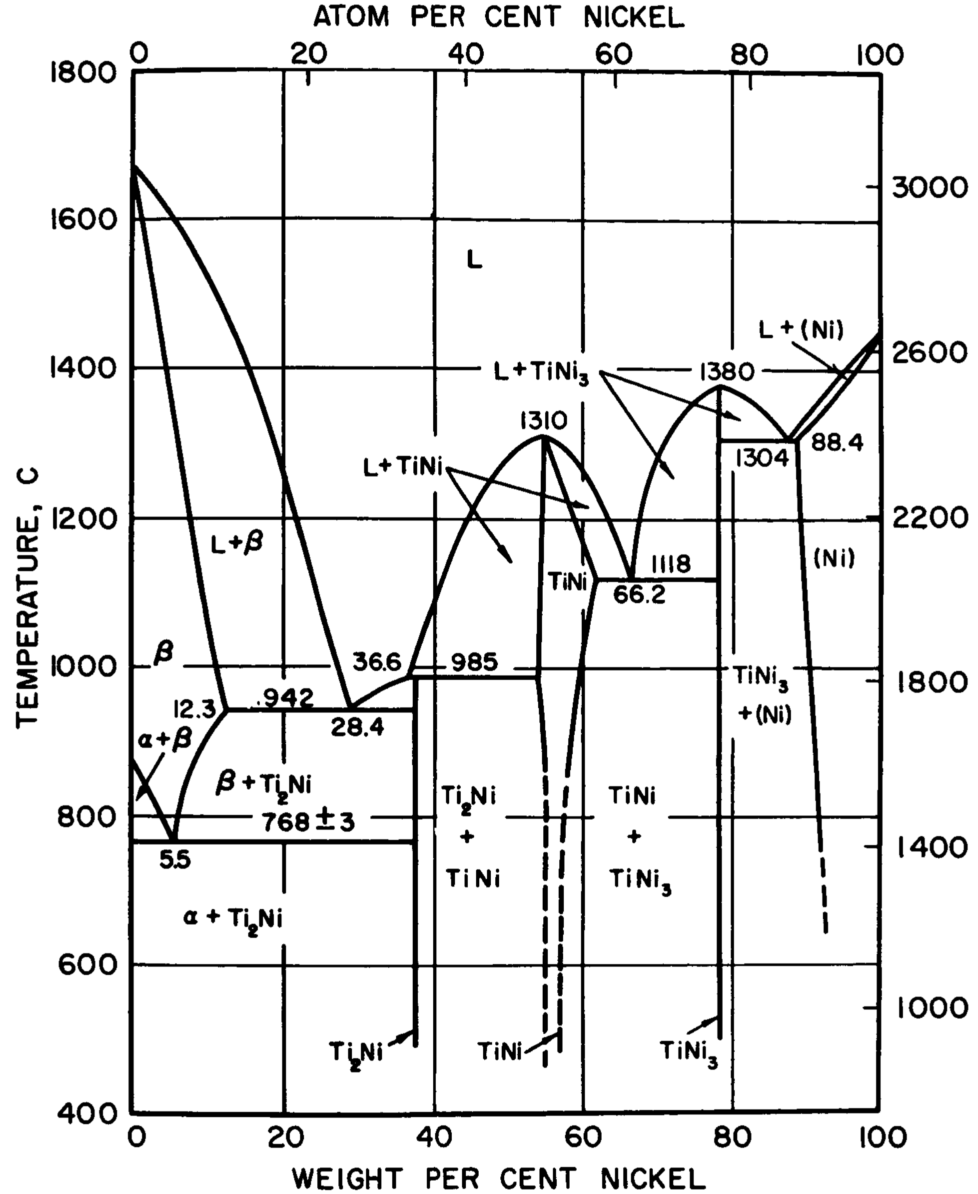

Figure 16 - Nickel-Titanium Phase Diagram

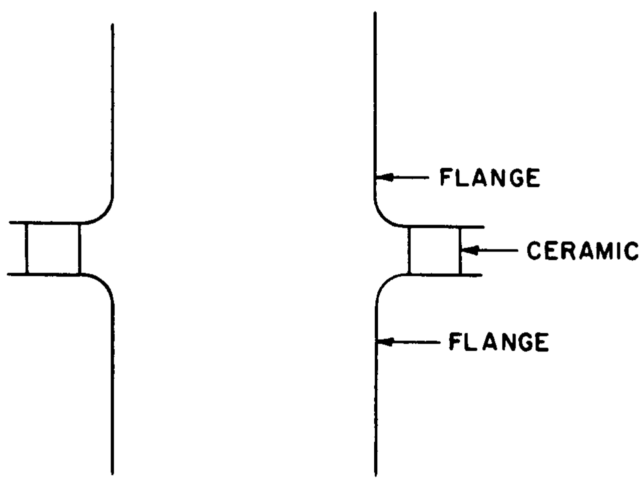

Figure 17 - Open Seal Structure

difficulties were avoided, the tube assembly was completed, and the tube is presently being exhausted.

3. Palladium-Cobalt Alloy

The palladium-cobalt alloy designated "Palco" could not be directly substituted for nickel titanium, since it did not wet the tantalum flanges. When a second tube was made with molybdenum flanges, wetting properties proved to be good and the tube was tight.

The interelectrode resistance in the new tube was greater than 20 megohms, compared to several hundred ohms for tubes made according to the design shown in Figure 1 with titanium-nickel alloy.

On the first high-temperature excursion, the tube resistance remained above 20 megohms up to temperatures of 400°C for the cathode and 600°C for the reservoir. Increasing the reservoir temperature to 650°C changed the apparent resistance to 2 megohms and, when the cathode temperature was then increased to 1100°C, the apparent tube resistance was approximately 1,000 ohms. It is not known whether there was actually a resistance path formed in the tube or whether the apparent tube impedance was low because of emission tendencies at the anode and cathode. After cooling, however, the tube impedance again registered about 20 megohms.

Testing is continuing on the Palco tube, Z-7009, No. 5, and more information should soon be available. This information should indicate the prospects not only of solving the halide-cycle riddle but also of selecting the right combination of electrode and reservoir temperatures to produce normal cathode emission.

The Z-7009, tube 4, which was similar to Z-7009, tube 1, contained titanium-nickel seals. When tube 4 was heated to approximately 750°C at the reservoir and 1400°C at the cathode, the tube impedance dropped to 0.1 ohm and remained at this value regardless of subsequent temperature state. It is suspected that the titanium-halogen cycle, previously discussed, was responsible for this condition. No cathode emission was observed at any time.

CESIUM IODIDE WITH CESIUM SEED

To date cathode emission, if occurring at all in CsI diodes, has been in the milliampere or microampere range. One hypothesis is that sufficient dissociation of CsI has not taken place to yield enough cesium to permit cesiated cathode emission. In the absence of a discharge, the CsI temperature has not exceeded the cathode temperature. If a discharge were formed, however, the CsI would be subjected to temperatures of several thousand degrees and thus be more likely to dissociate.

There is some interest in building a tube with a cesium iodide pellet (or infinite supply) and with cesium seeding, the amount of cesium being just sufficient to produce a few hundred microns of cesium pressure when fully vaporized. Such a tube should be capable of starting with the cesium seeding, with the arc discharge presenting a hitherto unavailable background condition for dissociation of CsI.

If such a tube is built, it will be constructed after the results of present experiments designed to eliminate the halogen cycle are known.

VAPOR FILL AND CATHODE MATERIALS

The present effort is concentrated on cesium-filled tubes with cesium-enhanced cathodes. It is felt that this is the most efficient combination in terms of power loss, because of the low work function of a cesiated cathode and the low ionization potential of cesium. It may also be the combination least likely to succeed in a high-temperature tube. In review, eight combinations of fill and cathode are listed in order of descending efficiency:

	Cathode	Fill
1.	Cesiated refractory metal	Cesium (high pressure)
2.	Barium	Cesium (low pressure)
3.	Barium	Thallium or equivalent
4.	Thoriated tungsten	Cesium (low pressure)
5.	Thoriated tungsten	Thallium or equivalent
6.	Tungsten	Cesium (low pressure)
7.	Tungsten	Thallium or equivalent

It is felt that the likelihood of succeeding in the design of a high-temperature tube increases as one moves down the list. For example, imagine two curves having opposite slopes, a descending "efficiency" curve and an increasing "likelihood" curve. The position at which these curves intersect will determine the best combination of cathode and fill. Work will continue with the most efficient combination until voltage and temperature limitations are definitely established.

THALLIUM TUBES

Two thallium tubes with barium cathodes are under construction. These tubes will not only permit an appraisal of a promising vapor, but will also provide advance data should subsequent test results indicate the necessity of selecting a lower efficiency combination of fill and cathode material.

One tube will be made in the diode structure of Figure 1, while the other will utilize the design of the high-temperature gas tube developed under Contract No. NAS3-2548 and displayed in Figure 18.

FIFTEEN-AMPERE THYRATRON DESIGN

It is impossible to formulate a final design of a thyratron before it is even known what combination of cathode and fill will be used. A conceptual design, however, is shown in Figure 19 for the following combinations:

	Cathode	Fill
1.	Cesium	Cesium
2.	Barium	Cesium
3.	Barium	Thallium or equivalent

Over-all length and diameter (including convectors) are approximately three inches and four inches, respectively. The grid and anode convectors will be securely bolted to the 600°C heat sink using the necessary beryllia wafers for voltage insulation. The heater element consists of a coaxial structure with the outer sheath brazed directly to the cathode surface.

It should be emphasized that the conceptual design of the thyratron is subject to change as more test data becomes available.

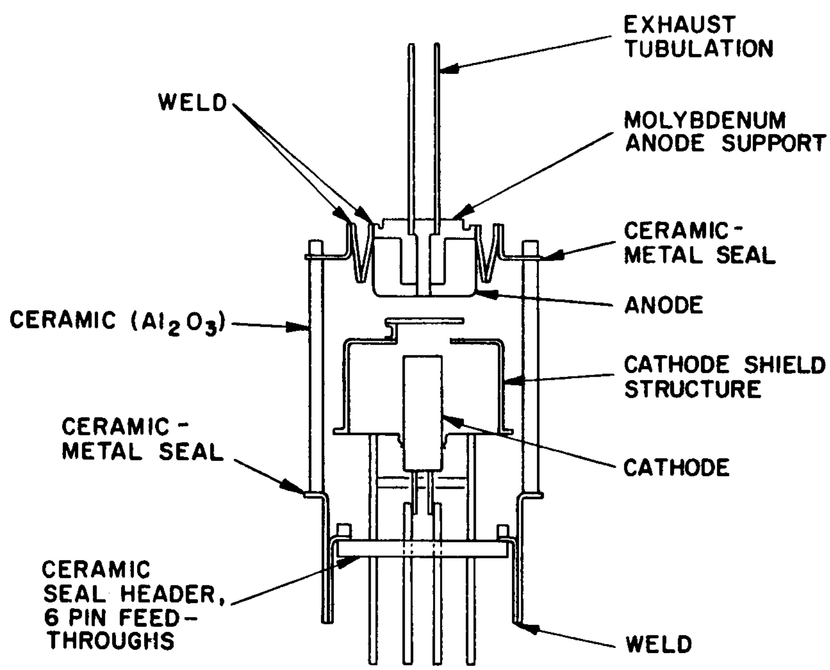

Figure 18 - Schematic of Test Vehicle, Design C

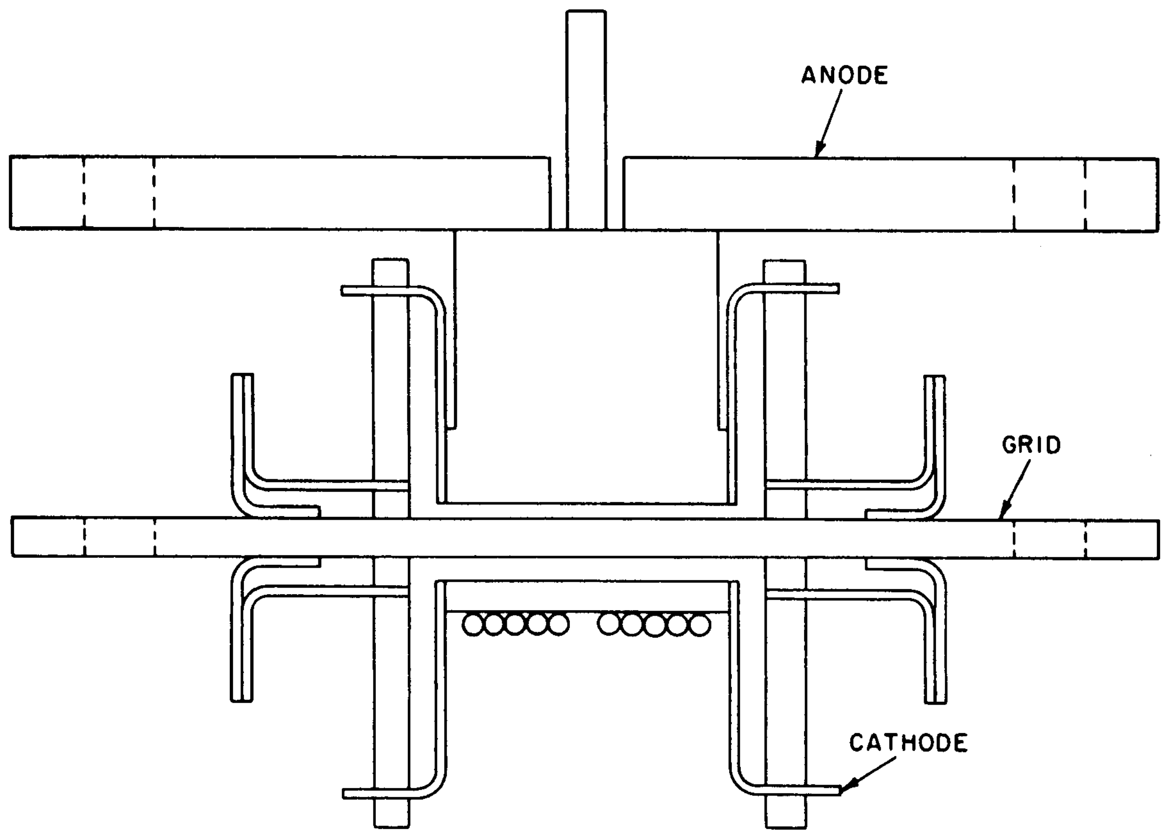

Figure 19 - Conceptual Design for High-Temperature Thyratron

PROGRAM FOR NEXT REPORT PERIOD

During the next report period, diodes having the following special features will be assembled, exhausted and tested:

1. Titanium-nickel brazing alloy covered with nickel plate
2. Zirconium-carbide coated anode
3. Hafnium coated anode
4. Long ceramic and small interelectrode gaps for low leakage and high voltage
5. Barium cathode, thallium fill, test diode structure
6. Barium cathode, thallium fill, gas tube structure
7. Cesium iodide with cesium seed (tentative).

In addition, further test data will be obtained on Z-7009, tube 5 with Palco seals. Also, the design for 15-ampere thyratron will be refined.

Development of

HIGH-TEMPERATURE, HIGH-CURRENT, ALKALI-METAL VAPOR-FILLED CERAMIC THYRATRONS AND RECTIFIERS

by

A. W. Coolidge

ABSTRACT

Cathode emission characteristics were obtained in two test diodes containing a pure cesium fill. Inverse emission characteristics of the anode were also measured and effort was expended in obtaining an anode surface from which inverse emission should be considerably reduced.

Work was continued in the area of seal redesign in an attempt to combat a bothersome halide cycle.

The test diode was redesigned in order to increase anode voltage ability.

Two tubes were started that will be filled with thallium and will have barium system cathodes.

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